

Origen de las propiedades magnetoeléctricas del Y_2CoMnO_6

II Congreso de
‘Materiales para los retos de la sociedad’

Javier Blasco Carral

Trabajo realizado por:

Caracterización de materiales mediante Técnicas de radiación de sincrotrón



Joaquín García Gloria Subías J. Blasco



Jolanta Stankiewicz

En colaboración con:



Alberto Rodríguez-Velamazán
Clemens Ritter



José Luis García-Muñoz



François Fauth



Esquema

✓ *Introducción:*

Estructura y magnetismo de dobles perovskitas

Materiales multiferroicos: Definición y clasificación

Antecedentes en el estudio del Y_2CoMnO_6

✓ *Propiedades del Y_2CoMnO_6 :*

Estructura cristalográfica y magnética.

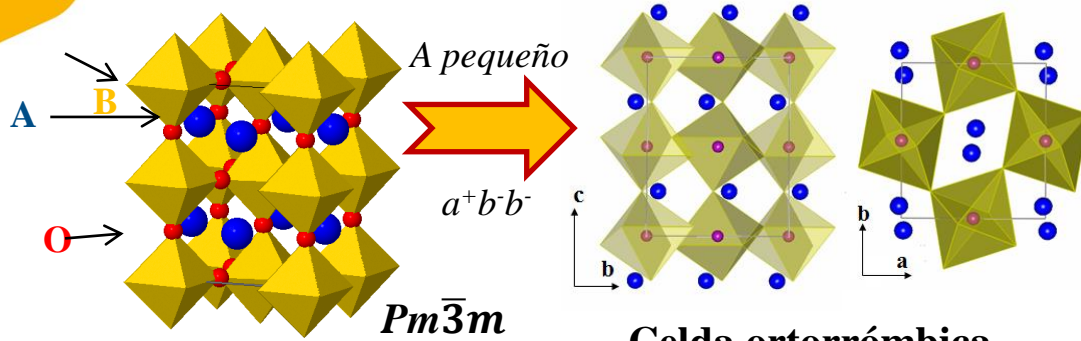
Propiedades magnéticas

Propiedades eléctricas

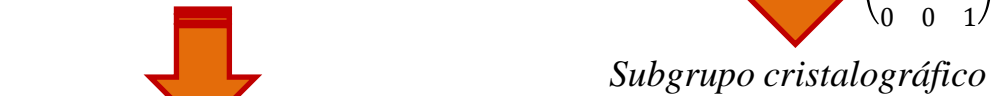
✓ *Conclusiones.*

Introducción: estructura y magnetismo

✓ *Cristalográfica*



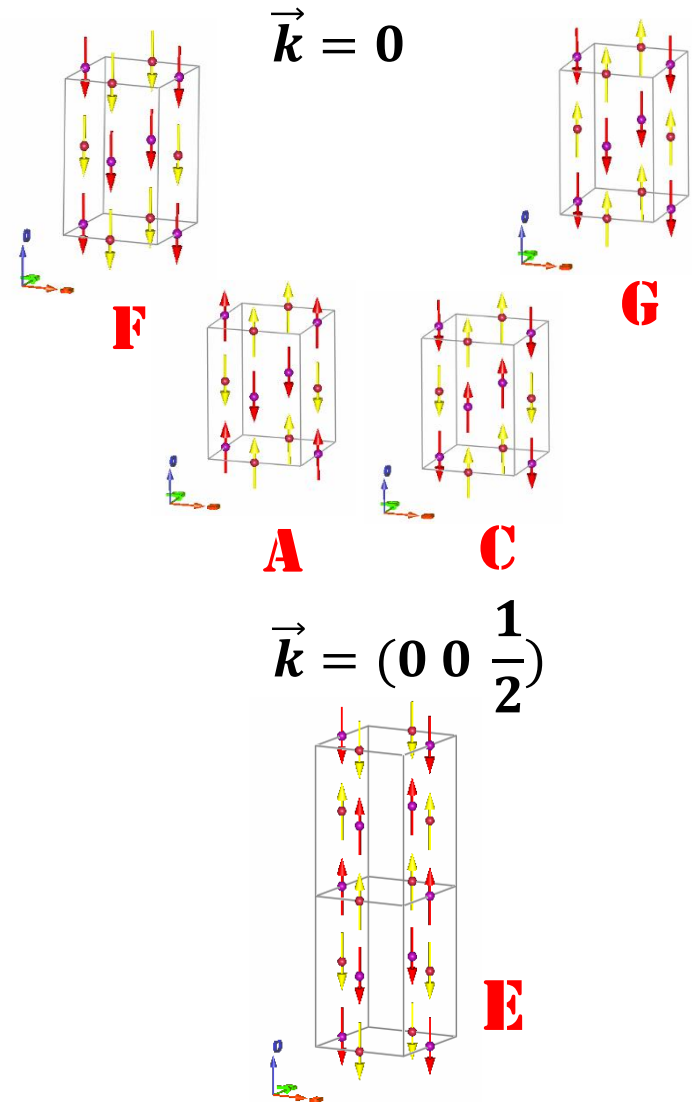
B diferentes (tamaño, carga)



Antisite defects (ASD) {

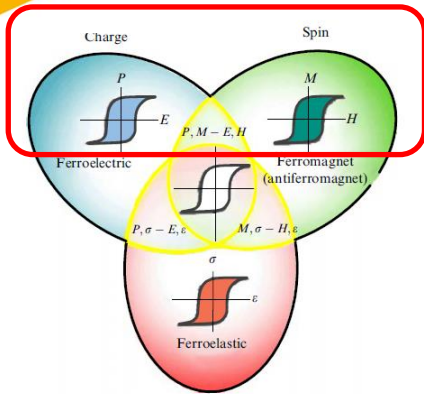
- 0 % → perovskita doble perfecta
- 50 % → perovskita simple

✓ *Magnética*



Introducción: Materiales multiferroicos

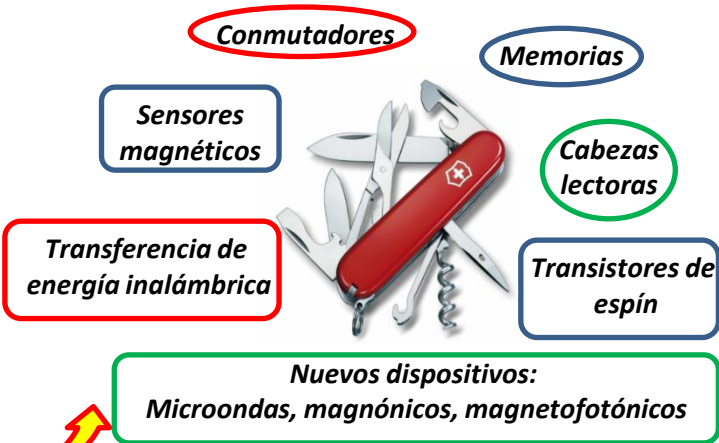
✓ Coexistencia de al menos 2 fases ferro



Multiferroicos magnetoeléctricos:

☝ Coexistencia de orden:
eléctrico y magnético

✌ Acoplamiento entre ambas propiedades:
acoplamiento magnetoeléctrico

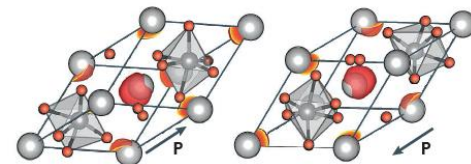


Nuevos materiales multifuncionales

✓ Clasificación de materiales multiferroicos magnetoeléctricos:

📁 **Tipo I:** Magnetismo y ferroelectricidad (FE) tienen diferentes orígenes.

- Temperaturas de transición muy diferentes (altas) 😊
- Débil acoplamiento magnetoeléctrico 😞

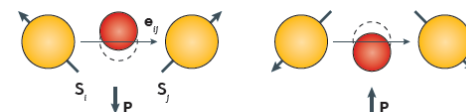


$T_C = 1103 \text{ K}$

$T_N = 643 \text{ K}$

📁 **Tipo II:** El magnetismo causa la FE.

- Idénticas temperaturas de transición (bajas) 😞
- Fuerte acoplamiento magnetoeléctrico 😊



$T_C = 29 \text{ K}$

Introducción: Materiales multiferroicos

Orden tipo E



Multiferroico tipo II

✓ Cálculos *ab-initio* proponen la estabilización del orden magnético de tipo E en perovskitas simples y dobles con A^{3+} pequeño ($A^{3+}=R^{3+}$ pesada o Y^{3+}).

✓ Acoplado a dicho orden se produce polarización eléctrica (P) perpendicular a la dirección del orden magnético.

✓ Dos ingredientes básicos:

Interacciones magnéticas competitivas (FM-NN + AFM-NNN)

Desplazamientos de los O acoplados al giro de octaedros BO_6 .

✓ Ejemplo en perovskita simple:

fase ortorrómbica del HoMnO_3 ($T_c=26\text{K}$) e YMnO_3 ($T_c=28\text{K}$)

IOP PUBLISHING

J. Phys.: Condens. Matter 20 (2008) 434208 (10pp)

JOURNAL OF PHYSICS: CONDENSED MATTER

doi:10.1088/0953-8984/20/43/434208

Microscopic mechanisms for improper ferroelectricity in multiferroic perovskites: a theoretical review

Silvia Picozzi¹, Kunihiro Yamauchi¹, Ivan A Sergienko^{2,3},
Cengiz Sen^{2,3,5}, Biplob Sanyal¹ and Elbio Dagotto^{2,3}

PHYSICAL REVIEW B 82, 134429 (2010)

Theoretical prediction of multiferroicity in double perovskite Y_2NiMnO_6

Sanjeev Kumar,^{1,2,3} Gianluca Giovannetti,⁴ Jeroen van den Brink,³ and Silvia Picozzi⁴

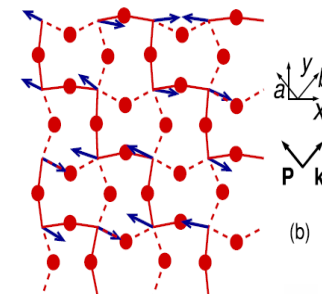
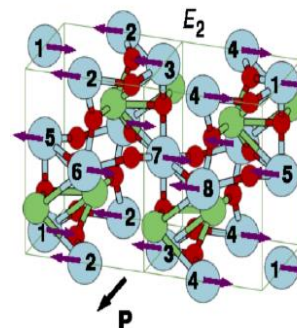
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(Received 13 November 2009; revised manuscript received 3 May 2010; published 18 October 2010)



Introducción: Antecedentes Y_2CoMnO_6

Multiferroic behavior in the new double-perovskite $\text{Lu}_2\text{MnCoO}_6$

S. Yáñez-Vilar¹, E. D. Mun², V. S. Zapf², B. G. Ueland³, J. Gardner^{4,5}, J. D. Thompson³, J. Singleton², M. Sánchez-Andújar¹, J. Mira⁶, N. Biskup⁷, M. A. Seánar-Rodríguez¹, C. D. Batista⁸

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⁷Dpto. Tecnologías de la Información Inst. de Ciencia de Materiales 28040 Madrid (Spain)

⁸Theory division, LANL, Los Alamos, NM 87545 (USA)

(Dated: May 12, 2011)

We present a new member of the multiferroic oxides, $\text{Lu}_2\text{MnCoO}_6$, which we have investigated using X-ray diffraction, neutron diffraction, specific heat, magnetization, electric polarization, and dielectric constant measurements. This material possesses an electric polarization strongly coupled to a net magnetization below 35 K, despite the antiferromagnetic ordering of the $S = 3/2$ Mn^{4+} and Co^{2+} spins in an $\uparrow\downarrow\downarrow$ configuration along the c -direction. We discuss the magnetic order in terms of a condensation of domain boundaries between $\uparrow\uparrow$ and $\downarrow\downarrow$ ferromagnetic domains, with each domain boundary producing a net electric polarization due to spatial inversion symmetry breaking. In an applied magnetic field the domain boundaries slide, controlling the size of the net magnetization, electric polarization, and magnetoelectric coupling.

APPLIED PHYSICS LETTERS 107, 012902 (2015)



Evidence of large magneto-dielectric effect coupled to a metamagnetic transition in $\text{Yb}_2\text{CoMnO}_6$

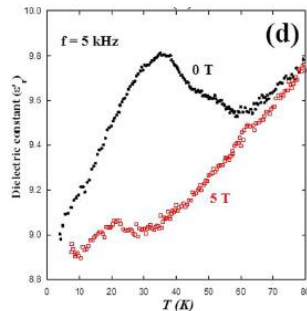
J. Blasco,^{1,a)} J. L. García-Muñoz,² J. García,¹ J. Stankiewicz,¹ G. Subías,¹ C. Ritter,³ and J. A. Rodríguez-Velamazán^{1,3}

¹Instituto de Ciencia de Materiales de Aragón, CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain

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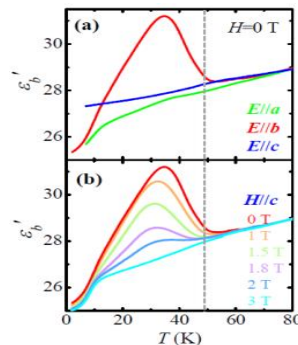
✓ Aumento de la casuística:
 $\text{Yb}_2\text{CoMnO}_6$ adopta el orden E

✓ Efecto magnetocapacitivo explicado
por transición metamagnética $E \rightarrow F$

✓ No hay FE ¿Defectos?

✓ Primer hallazgo del orden E en una doble perovskita (eje c).

✓ Anomalía en ϵ' acoplada al orden magnético dependiente de H_{ext}



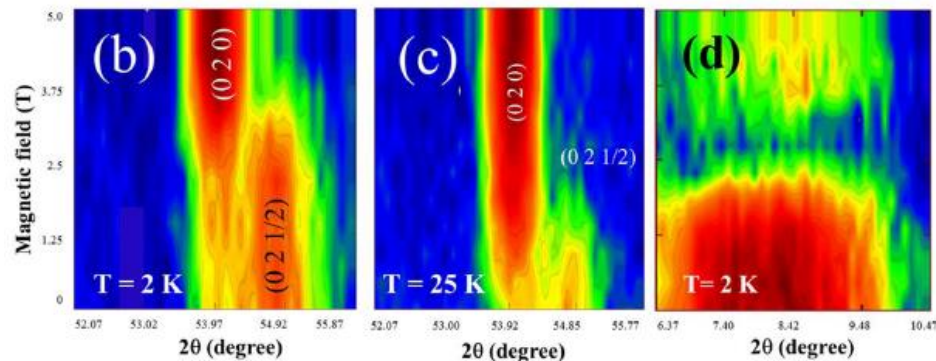
Estudio de Monocrystal:

N. Lee et al., APL 104, 112907 (2014)

✓ Comportamiento anisotrópico de ϵ'

✓ Confirma efecto magnetocapacitivo

✓ No se observa FE



Y_2CoMnO_6 : Antecedentes

2013

APPLIED PHYSICS LETTERS 103, 012603 (2013)



Magnetism driven ferroelectricity above liquid nitrogen temperature in Y_2CoMnO_6

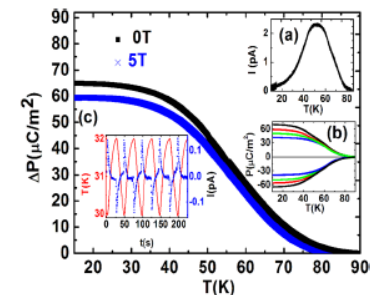
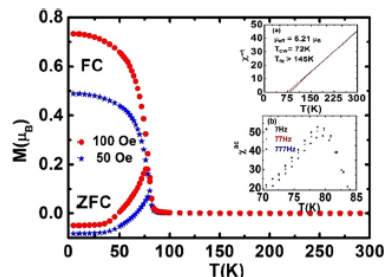
G. Sharma,¹ J. Saha,¹ S. D. Kausik,² V. Srugan,² and S. Patra^{1,*}
¹School of Physical Sciences, Jawahar Institute of Physics, New Delhi 110029, India
²UGC-DARE Consortium for Scientific Research, Mumbai Centre, R. S. Shinde, BARC, Mumbai 400085, India

(Received 28 February 2013; accepted 16 June 2013; published online 3 July 2013)

We report multiferroic behaviour in double perovskite Y_2CoMnO_6 with ferroelectric transition temperature $T_f = 80$ K. Both X-ray diffraction and neutron scattering data confirm a centrosymmetric crystal structure of space group $P2_1/a$ at room temperature. The saturation polarization and magnetization are estimated to be $65 \mu\text{C}/\text{m}^2$ and $6.2 \mu_B/\text{f.u.}$, respectively. The magneto-electric coupling parameter, on the other hand, is small as a 5 T field suppresses the electric polarization by only ~8%. The origin of ferroelectricity is associated with magnetic ordering of Co^{2+} and Mn^{4+} moments in $[\text{T}-1]$ arrangement. A model based on exchange-striction is proposed to explain the observed high temperature ferroelectricity. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4812728]

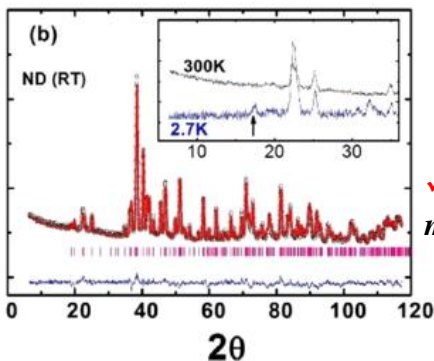
✓ Efecto piroeléctrico a similar temperatura que el orden magnético.

✓ Acoplo magnetoeléctrico !



✓ Detectan un pico magnético no F.

✓ Presuponen un orden E. No muestran ajuste.



2015



PCCP

PAPER



Chem. Mater. Phys. Chem. Phys., 2015, 17, 2094

The ferroelectric polarization of Y_2CoMnO_6 aligns along the b-axis: the first-principles calculations

C. Y. Ma,^{1,2} Dong¹ P. X. Zhou,¹ Z. Q. Du,¹ M. F. Liu,¹ H. M. Liu,¹ Z. B. Yan¹ and J.-M. Liu^{1*}

Double perovskite AB_2O_6 oxides with magnetic B and B' ions and E-type antiferromagnetic order (E-AFM, i.e. the $[\text{T}-1]$ structure) are believed to exhibit promising multiferroic properties, and Y_2CoMnO_6 (YCMO) is one candidate in this category. However, the microscopic origins for magnetically induced ferroelectricity in YCMO remain unclear. In this study, we perform detailed symmetry analysis on the exchange-striction effect and lattice distortion plus the first-principles calculations on YCMO. The E-AFM state as the ground state with other competing states such as ferromagnetic and A-antiferromagnetic orders is confirmed. It is observed that the ferroelectricity is generated by the exchange-striction associated with the E-AFM order and chemically ordered MnCo₂ occupation. Both the lattice symmetry consideration and first-principles calculations predict that the electric polarization aligns along the b-axis. The calculated polarization reaches up to $5.46 \times 10^{-3} \text{ C}/\text{cm}^2$, mainly from the spin displacement contribution. The present study presents a comprehensive understanding of the multiferroic mechanisms in YCMO and is of general significance for predicting emergent multiferroicity in other double perovskite magnetic oxides.

✓ Cálculos: Fase E más estable que F o A (fuerte competencia)

✓ P alineada con eje b y un valor de $\sim 0.5 \mu\text{C}/\text{cm}^2$ (~100 veces mayor que el dato experimental)



PERO !!!

JOURNAL OF APPLIED PHYSICS

VOLUME 88, NUMBER 1

1 JULY 2000

Magnetic phase diagrams of the $\text{Ln}(\text{Mn}_{1-x}\text{Co}_x)\text{O}_3$ ($\text{Ln}=\text{Eu}, \text{Nd}, \text{Y}$) systems

I. O. Tsvetkov¹ and D. D. Kholapov²
¹Institute of Solid State and Semiconductor Physics, National Academy of Sciences of Belarus, P. Bruck St. 11, 220027 Minsk, Belarus

J. W. Lynn and R. W. Erwin

NIST Center for Materials Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899-8242

Q. Huang

Department of Materials and Nuclear Engineering, University of Maryland, College Park, Maryland 20742

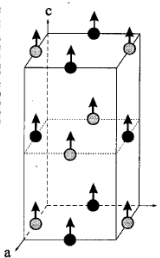
H. Szymczak, R. Szymczak, and M. Baran

Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Wł.

(Received 9 September 1999; accepted for publication 4 February 2000)

We report magnetization measurements in the $\text{Ln}(\text{Mn}_{1-x}\text{Co}_x)\text{O}_3$ ($\text{Ln}=\text{Eu}, \text{Nd}, \text{Y}$) systems.

Compositions in the range of $0.35 \leq x \leq 0.8$ ($\text{La}=\text{Eu}$), $0.4 \leq x \leq 0.6$ ($\text{La}=\text{Nd}$) exhibit antiferromagnetic behavior associated with a transformation from ferromagnetic, where the magnetic moments of Co^{2+} and Mn^{2+} ferromagnetic spins are parallel. A ferromagnetic state is observed ($\text{La}=\text{Nd}$) and $0.45 \leq x \leq 0.5$ ($\text{La}=\text{Y}$) compositions. Doping and $H-T$ phase diagrams resemble those of $\text{La}_2\text{CoMnO}_6$ charge-order diffusion studies indicate the order of the Co and Mn ions in the which is presumably responsible for its ferromagnetic behavior. © 2000 AIP Publishing LLC. [S0021-8995/00/0101-01]



Available online at www.sciencedirect.com

ScienceDirect

Progress in Solid State Chemistry 35 (2007) 257–264



www.elsevier.com/locate/psch

Neutron diffraction evidence for a cationic order in the $\text{REMn}_{0.5}\text{Ni}_{0.5}\text{O}_3$ ($\text{RE}=\text{La}, \text{Nd}$) and $\text{YMn}_{0.5}\text{Co}_{0.5}\text{O}_3$ perovskites

M. Mouallem-Bahout^{a,*}, T. Roisnel^a, F. Bourée^b, G. André^b, C. Moure^c, O. Peña^a

JOURNAL OF APPLIED PHYSICS 106, 123607 (2009)



Magnetization steps in Y_2CoMnO_6 double perovskite: The role of antisite disorder

H. Szymczak,¹ B. Nijm,¹ R. Szymczak,² V. V. Kabanov,³ D. Chertan,⁴ S. Bujak,⁵ Thomas Hansen,⁶ J. P. Chaudhary,⁷ and Th. Brückel¹

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(Received 1 August 2014; accepted 12 September 2014; published online 12 September 2014)

Antisite disorder is observed to have significant impact on the magnetic properties of double perovskites Y_2CoMnO_6 , which has been recently identified as a ferromagnetic phase transition occurs in this material at $T_f = 75$ K.

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✓ FM en el eje c.

✓ Muestra con vacantes catiónicas.

✓ Estequiometría Co/Mn ~ 1

✓ FM perpendicular al plano ab.

✓ Muestra con gran desorden

ASD ~ 34%

✓ FM en el eje c con 'canting'.

✓ Ajuste con dos fases:.

- 38% Ortorrómica sin ordenar

- 62% monoclinica con ASD ~ 30%

✓ Fuertes contradicciones entre trabajos.

✓ ¿Diferentes muestras?

¿Cuál es la estructura magnética del Y_2CoMnO_6 con la estequiometría adecuada? ¿Y sus propiedades?

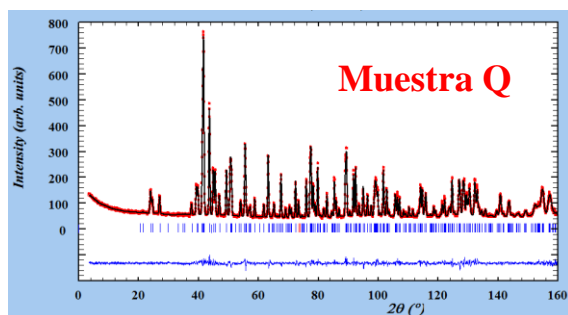
Y_2CoMnO_6 : Muestras y estructura

✓ Preparación de 3 muestras:

1. Muestra Q: $1250^\circ\text{C} \rightarrow$ 'Quench' al aire
2. Muestra M: $1250^\circ\text{C} \rightarrow 300^\circ\text{C}$ a $5^\circ\text{C}/\text{min}$
3. Muestra S: $1250^\circ\text{C} \rightarrow 300^\circ\text{C}$ a $0.1^\circ\text{C}/\text{min}$

✓ Composición química similar:

Element	Nominal	Q-sample	M-sample	S-sample
Y	60.96	60.84 ± 0.26	60.84 ± 0.26	60.70 ± 0.26
Co	20.20	20.15 ± 0.19	20.17 ± 0.19	20.26 ± 0.19
Mn	18.84	19.01 ± 0.20	18.99 ± 0.20	19.04 ± 0.20



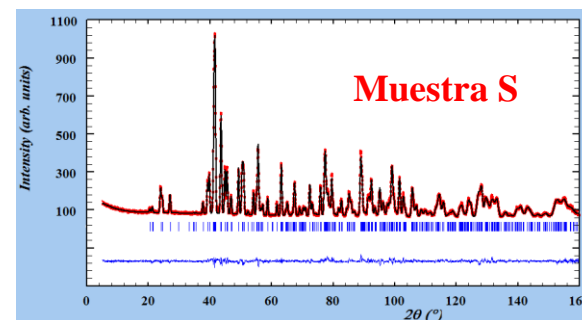
✓ Difracción de neutrones a 300 K

Determinación de ASD:

Muestra Q = 46.3(17)%

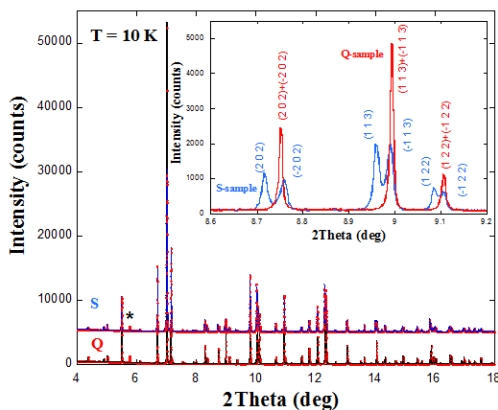
Muestra M = 21.1(9)%

Muestra S = 8.2(6) %



✓ Distancias interatómicas $\rightarrow \text{Mn}^{4+}-\text{O}$ y $\text{Co}^{2+}-\text{O}$

✓ Homogeneidad de las muestras analizadas mediante XRD de alta resolución (HRXRD): Línea MSPD en ALBA-CELLS

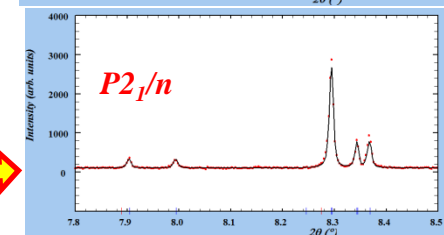
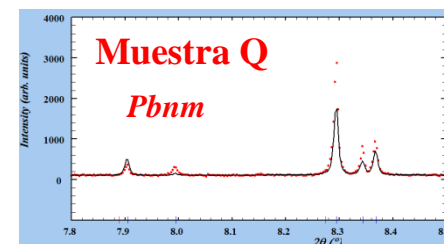


Picos estrechos \rightarrow resolución instrumental

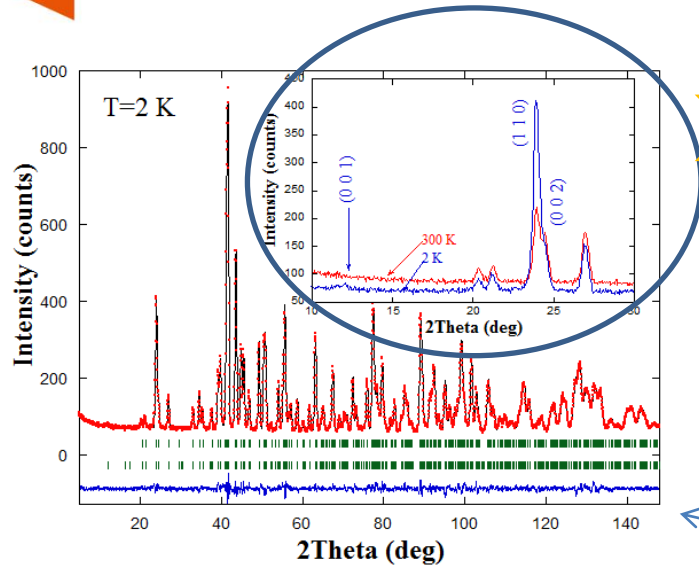
Disminución de la distorsión monoclinica al aumentar ASD:

Muestra Q $\rightarrow \beta \sim 90^\circ$ (89.984°)

HRXRD determina que todas las muestras son monoclinicas



Y_2CoMnO_6 : Estructura magnética



✓ La comparación de los difractogramas de neutrones muestra:

1. Todos los picos magnéticos siguen el vector $\vec{k} = 0$
2. Fuerte contribución FM en el eje c
3. Contribución FM en el plano ab.
4. Débil componente AFM en el plano ab

No hay Fase E

✓ *Análisis de Simetría:*

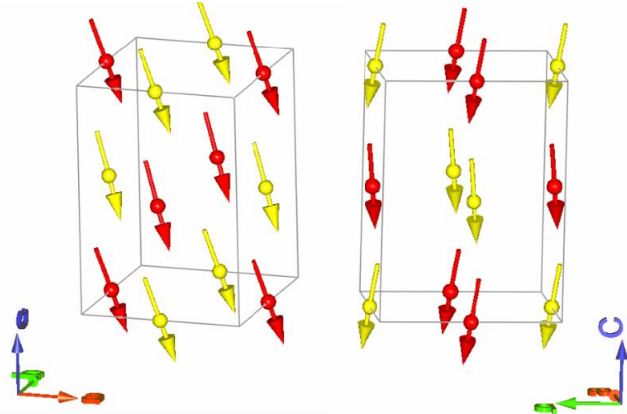
Base vectors for sites (2d) and (2c)							
Γ_v	1	2_y	Co(1)	Co(2)	Mn(1)	Mn(2)	Orden
Γ_1	1	1	(100) (010) (001)	(-100) (010) (00-1)	(100) (010) (001)	(-100) (010) (00-1)	A_x F_y F_z
Γ_3	1	-1	(100) (010) (001)	(100) (0-10) (001)	(100) (010) (001)	(100) (0-10) (001)	F_x A_y F_z

Signo opuesto

C_x
 G_y
 C_z
 G_x
 C_y
 G_z

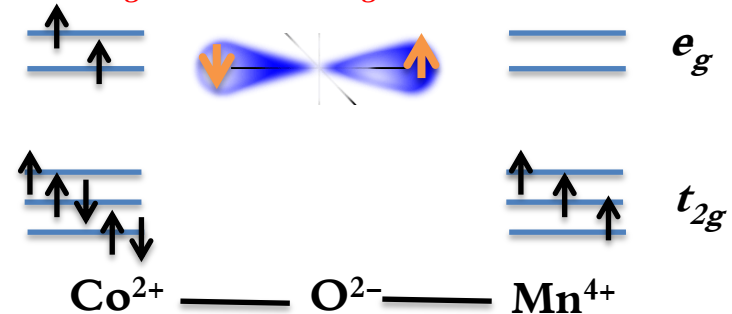
✓ Ajuste estable: $\mu_{\text{Co}} = \mu_{\text{Mn}} \Rightarrow \text{Co}^{2+} \text{ y } \text{Mn}^{4+} \text{ (HS)} \Rightarrow 3\mu_B/\text{at.}$

✓ Ajuste $\mu_t = (0.85, 0.33, 2.78)$; $|\mu_t| = 2.92(2) \mu_B/\text{at.}$



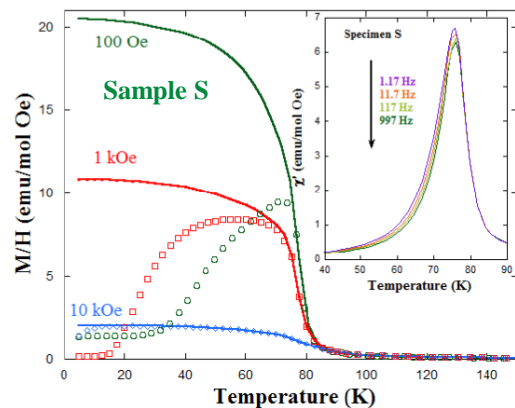
$\text{Co}(1) = \frac{1}{2} 0 0$; $\text{Co}(2) = 0 \frac{1}{2} \frac{1}{2}$; $\text{Mn}(1) = \frac{1}{2} 0 0$; $\text{Mn}(2) = \frac{1}{2} 0 \frac{1}{2}$

Reglas de Goodenough-Kanamori



Y_2CoMnO_6 : propiedades magnéticas

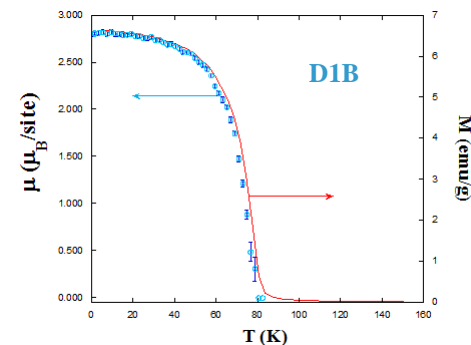
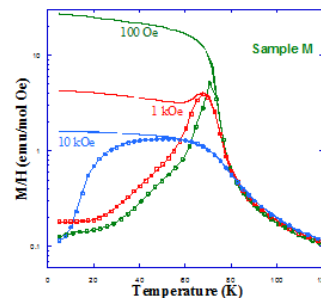
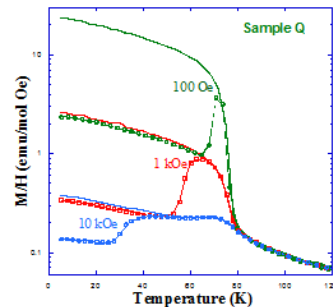
✓ Irreversibilidad ZFC-FC



✓ Las curvas de imanación dependen de la historia magnética de la muestra.

✓ Ligera dinámica en la transición.

✓ Contribución de dominios magnéticos anclados.



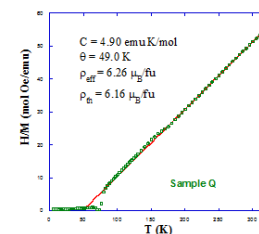
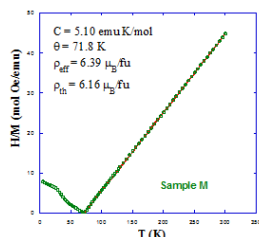
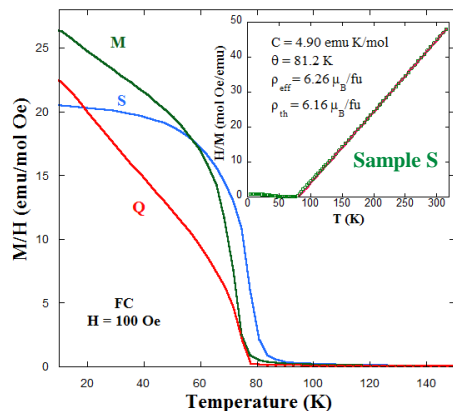
✓ La evolución en temperatura de los momentos refinados por neutrones mimetiza la curva de imanación.

✓ $T_C \sim 78$ K

✓ Efecto de los ASD en $M(T)$

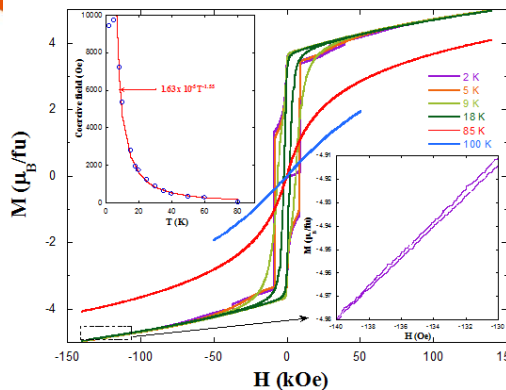
✓ T_C depende poco de los ASD

✓ $T > T_C$ sigue la ley de Curie-Weiss con interacciones FM

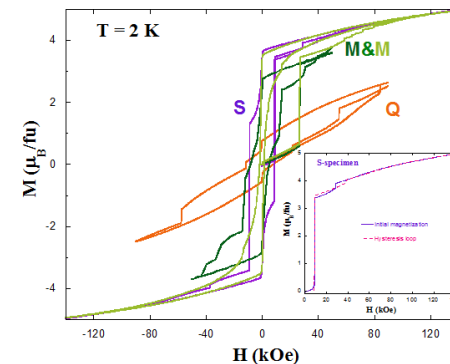


Specimen	Cooling conditions	ASD (%)	C (emu K/mol)	θ (K)	ρ_{eff} (μ_B/fu)
S-sample	0.1°/min	8.2	4.90	81.2	6.26
M-sample	5°/min	21.1	5.10	71.8	6.39
Q-sample	quench	46.3	4.90	49.0	6.26

Y_2CoMnO_6 : propiedades magnéticas



- ✓ Imanación espontánea a $T < T_C$.
- ✓ $H_C \propto T^{3/2}$ para $T > 8$ K. Expansión de los ejes donde se ordenan los momentos.
- ✓ No hay saturación para $\mu_0 H = 14$ T.
- ✓ A bajos $H \longrightarrow M \propto ASD$.
- ✓ Saltos evidencian avalanchas magnéticas.

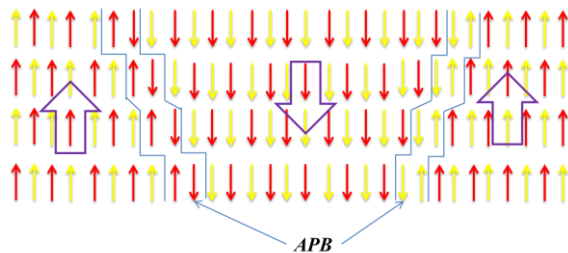


Hipótesis: Existencia de 'antiphase boundaries'

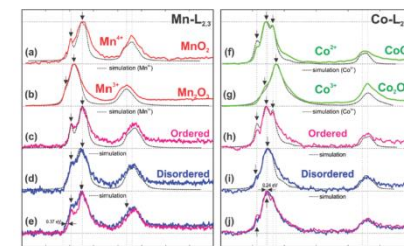
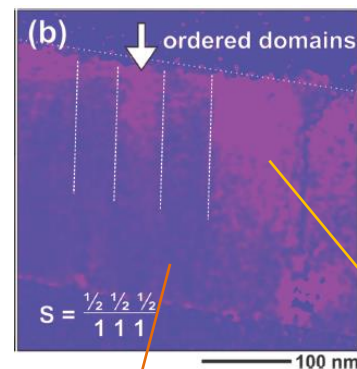
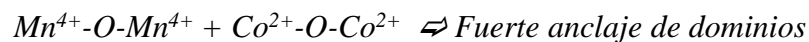
Hallados en sistemas similares: $\text{La}_2\text{CoMnO}_6$

Nanoscale, 2015, 7, 9835

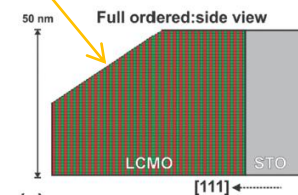
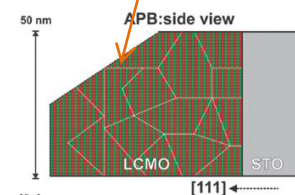
ASD $\left\{ \begin{array}{l} d=1 \Leftrightarrow \text{Defecto puntual} \\ d=2, 3 \Leftrightarrow \text{APB} \end{array} \right.$



Interacciones de superintercambio AFM:

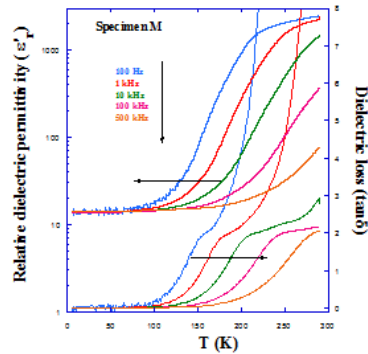


✓ Regiones ordenadas y desordenadas con las mismas especies iónicas.



Y_2CoMnO_6 : propiedades eléctricas

✓ Permitividad dieléctrica

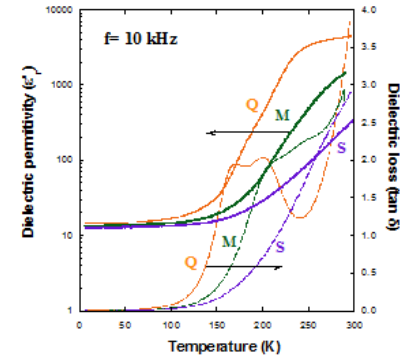
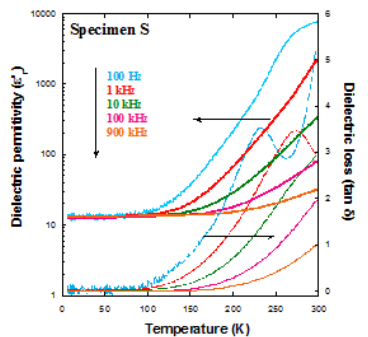
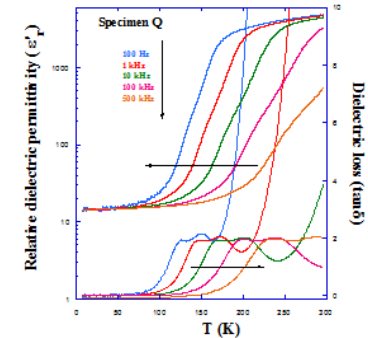


- ✓ ϵ_r' cte. a bajas temperaturas (~ 13).
- ✓ Saltos de ϵ_r' acoplados a pico en $\tan\delta$.
- ✓ Fuerte dependencia con la frecuencia



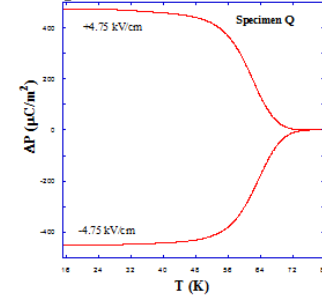
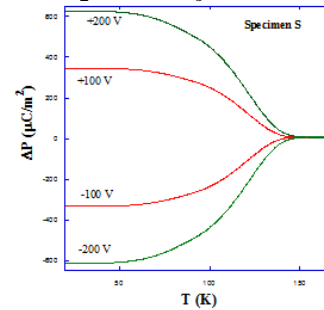
Mecanismos de barrera.

Contribuciones Maxwell-Wagner.

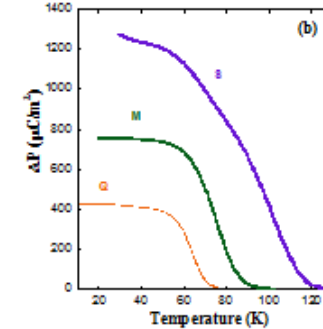
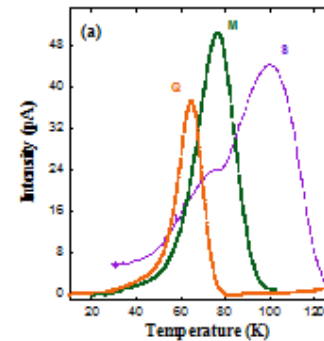


✓ Corriente piroeléctrica

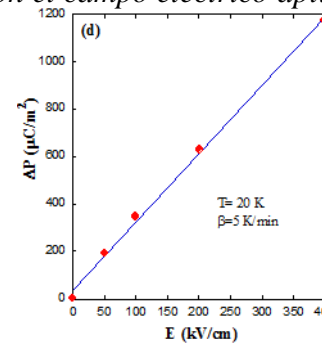
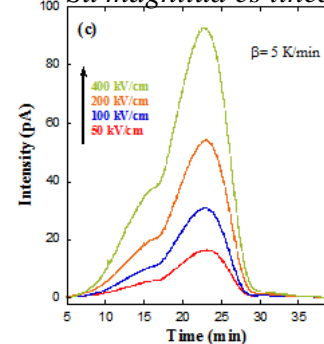
Y_2CoMnO_6 muestra efecto piroeléctrico reversible.



La corriente piroeléctrica depende del tipo de muestra.

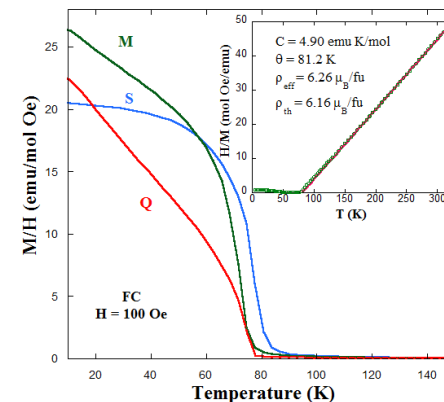
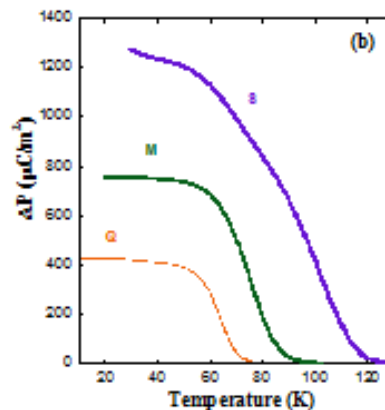


Su magnitud es lineal con el campo eléctrico aplicado.



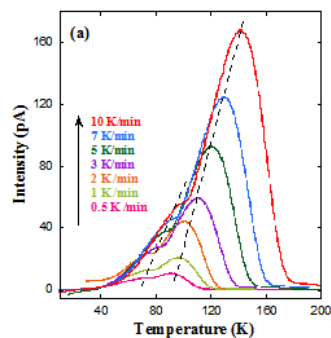
Y_2CoMnO_6 : efecto piroeléctrico

¿Transición ferroeléctrica en el Y_2CoMnO_6 ?
¿Acoplada al orden magnético?

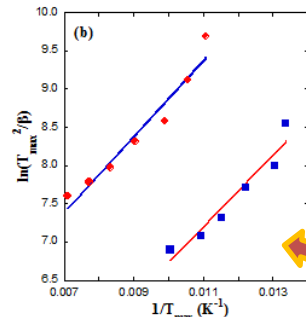


👉 *Diferente comportamiento de las dos transiciones. Acople casual!*

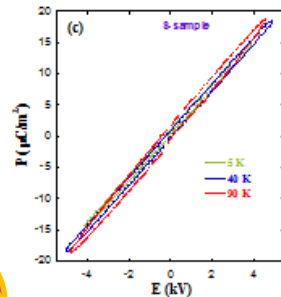
La corriente piroeléctrica depende de la dinámica de la medida!



Posición del pico sigue el modelo de Bucci



$$\frac{U}{k_B} = \frac{1}{\beta\tau^0} e^{\left(\frac{-U}{k_B T_{\max}}\right)} T_{\max}^2$$

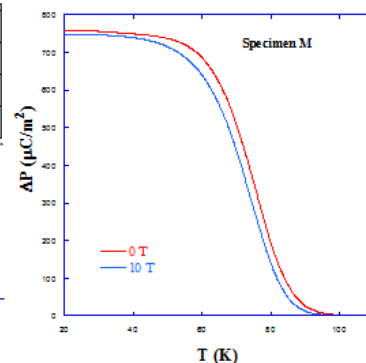
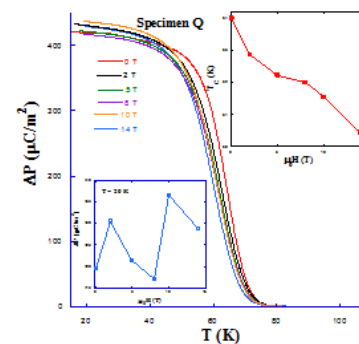


Ciclos de histéresis

Dieléctrico

Depolarización estimulada térmicamente \Leftrightarrow Dipolos asociados a defectos

✓ Acoplamiento magnetoeléctrico



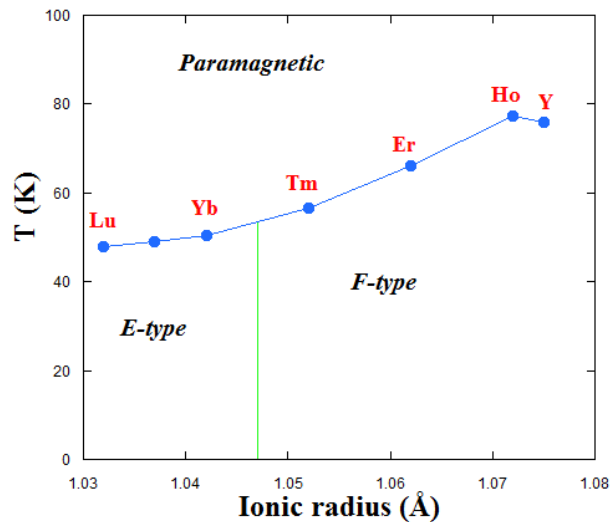
Despreciable

Orígenes diferentes para las polarizaciones eléctrica y magnética

CONCLUSIONES: Diagrama de fases



Estructuras tipo F y E compiten en R_2CoMnO_6 . Dependencia con r_R^{3+} :



✓ Fase F prevalece para $r_R^{3+} \geq 1.052$

✓ Fase E prevalece para $r_R^{3+} \leq 1.042$

✓ Y_2CoMnO_6 es un ferromagneto colineal.



\vec{P} asociada a defectos \Rightarrow No transición ferroeléctrica \Rightarrow No multiferroico.

¡Muchas Gracias
por
vuestra atención!